10^{-10} temporal contrast for femtosecond ultraintense lasers by cross-polarized wave generation

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We take advantage of nonlinear properties associated with $\chi^{(3)}$ tensor elements in BaF_2 cubic crystal to improve the temporal contrast of femtosecond laser pulses. The technique presented is based on cross-polarized wave (XPW) generation. We have obtained a transmission efficiency of 10% and 10^{-10} contrast with an input pulse in the millijoule range. This filter does not affect the spectral shape or the phase of the cleaned pulse. It also acts as an efficient spatial filter. In this method the contrast enhancement is limited only by the extinction ratio of the polarization discrimination device. © 2005 Optical Society of America

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One of the last major difficulties in the development of ultraintense and ultrashort laser systems is the ability to produce pulses with high temporal contrast. A typical high-power $Ti:Al_2O_3$ laser system, based on a chirped-pulse amplification (CPA) scheme, generates not only a femtosecond pulse but also an amplified spontaneous emission (ASE) nanosecond background as well as short prepulses and postpulses. To characterize the temporal quality of the pulse we define the incoherent pulse contrast by the intensity ratio between the ASE pedestal or satellite pulses and the main pulse. The temporal contrast for 100-TW class lasers currently reaches 6 orders of magnitude. For high-field physics experiments, the laser beam is focused with an intensity of 10^{21} W cm^{-2} on a solid target. This plasma-creating laser pulse is required to have high contrast to prevent the production of a preplasma before the main pulse reaches the target. Consequently, for laser–matter interaction applications it is crucial to decrease the pulse contrast to 10^{-9} or even better. To produce clean pulses it is essential to design new schemes for pulse amplification. Indeed, it has been reported that Kerr-lens mode-locked oscillators exhibit a very high-contrast (10^{-10}).[1] The ASE background is generated mostly in the preamplifier and then amplified with the main femtosecond pulse in power amplifiers, which produce less ASE.

Some techniques for improving temporal contrast, such as femtosecond preamplification[2] and the use of a saturable absorber[2,3] have been proposed. Also, it has been demonstrated that an efficient way to clean the pulse after preamplification is to set up a nonlinear filter, as was first illustrated by a nonlinear elliptical polarization rotation[4,5] (NER) in hollow waveguides filled with xenon, with a limited input energy of 100 μJ.[6] To increase the energy of the cleaned pulse, other schemes operating at the millijoule level have been proposed. These high-efficiency transmission filters are based on a nonlinear Sagnac interferometer[7,8] and on nonlinear polarization rotation in air.[9,10]

In this Letter we report on the production of 10^{-10} contrast pulses by use of a new nonlinear filtering technique based on cross-polarized wave (XPW) generation in nonlinear crystals.

XPW generation is a four-wave mixing process governed by the anisotropy of the real part of the crystal third-order nonlinearity tensor [$\chi^{(3)}$]. A full description of this process was previously detailed for cubic and tetragonal crystals.[11,12] The XPW generated wave has the same wavelength as the input pulse and a cubic dependence on the intensity. Consequently it potentially should facilitate an improvement in pulse temporal contrast.

As the medium is isotropic with respect to linear optical properties, the process is characterized by perfect phase and group-velocity matching of two orthogonally polarized waves propagating along the z axis. This property permits good efficiency of XPW generation and minimal pulse shape and spectral distortions. In our experiments we use a BaF_2 crystal ($m3m$ point group symmetry).

The XPW efficiency is proportional to the product of $\chi^{(3)}_{xxxx}$ and the anisotropy of the $\chi^{(3)}$ tensor[10] $\{\sigma = [\chi^{(3)}_{xxxx} - 2\chi^{(3)}_{xyyx} - \chi^{(3)}_{xxyy}]/\chi^{(3)}_{xxxx}\}$, when the self-phase modulation is determined mainly by $\chi^{(3)}_{xxxx}$. In our case the BaF_2 crystal has a $\chi^{(3)}_{xxxx}$ value that is moderate ($1.59\ 10^{-22}\ m^2/V^2$), but the anisotropy of $\chi^{(3)}$ is important ($\sigma=-1.2$).[11] These vales allow high-efficiency XPW generation but without excessive self-phase modulation.

Another advantage of BaF_2 is its transmission from the ultraviolet to the infrared. As its bandgap energy is high (9.07 eV), multiphoton absorption is negligible. So the $\chi^{(3)}$ value should be almost constant in the visible and the near infrared,[13] and XPW generation can be applied to various femtosecond laser wavelengths.

This promising system can also be adapted to other pulse energies by geometrical tuning, as the efficiency of the XPW generation is determined by the peak power intensity of the laser. We have demonstrated equal efficiency for input laser pulses from the microjoule to millijoule range.

Experiments were performed with a $Ti:Al_2O_3$ CPA laser including regenerative and multipass amplifiers. The laser system produces 42-fs, 2-mJ maximal energy pulses at a 1 kHz repetition rate. The input pulse is linearly polarized and focused by an $f'=3$ m lens. The BaF_2 crystal is 2 mm long and placed after the focal point to optimize the conversion process by reaching the correct peak intensity level ($\approx 3\times 10^{12}\ W\ cm^{-2}$). The crystal is rotated at an optimized angle,[11] β (the angle between the input polarization direction and its [100] axis). Then an analyzer transmits the XPW generated signal. The measured extinction ratio of the polarizer–analyzer pair is $\sim 5\times 10^{-5}$.

When the input energy was 1.2 mJ, we succeeded in producing cleaned pulses with an energy of 120 μJ and low self-phase modulation. Thus the uncorrected energy transmission efficiency of the setup was 10%. By taking into account the losses that are due to reflections on the crystal and analyzer interfaces, we can assert that an internal efficiency of better than 15% is achieved. This efficiency is still lower than efficiencies obtained by other techniques such as the use of NERs. In any case, the cleaned signal is energetic enough to be amplified into a second CPA setup. This might be a way to compensate for the energy lost in the filter, provided that the contrast is not excessively degraded during the second amplification.

We estimated the contrast improvement by using a homemade high-dynamic-range third-order cross correlator. We measured the filtered and nonfiltered pulses with the same energy seeding the correlator (120 μJ) to get comparable curves, as shown in Fig. 1. For this energy level the correlator noise is 10^{-11}. The ASE intensity level of the input pulse is 6 orders of magnitude below the peak intensity of the main pulse. As the XPW generated pulse intensity profile exhibits a cubic dependence on the input profile,[11,12] the pulse pedestal is drastically reduced. The 10^{-10} remaining ASE pedestal corresponds to a leakage of the incoming pulse pedestal through the analyzer. The extinction ratio of the polarizer–analyzer pair (5×10^{-5} in our experiment) corresponds to the recorded decrease of the ASE background. The intensity of parasitic pulses is reduced by the same amount, as the figure shows. For polarization discrimination techniques such as XPW generation and the use of NERs, the extinction ratio between polarizer and analyzer determines the overall contrast improvement. We believe that a XPW takes advantage of this property, as a NER requires the use of two quarter-wave plates between the polarizer and the analyzer, which degrade the extinction ratio.[4]

To summarize, the nonlinear filter that we designed leads to an improvement in temporal contrast by 4 orders of magnitude. The filtered pulse exhibits a contrast that reaches 10^{-10}. This performance is limited only by the extinction ratio of the polarizing elements.

We now study the spectral behavior of the XPW process. As the system is scalable in energy, we have characterized the same setup with a 150-μJ input pulse and an $f'=1$ m lens. In this case, measurements made with the same conversion efficiency revealed that the spectrum is not degraded by the conversion process, as shown in Fig. 2, which represents the nonfiltered and filtered spectra for a transmission efficiency of 10%. No modulation defect is visible, the spectral width is unchanged, and the spectrum exhibits a smooth Gaussian shape. As the beam is focused in air, at higher input energy levels self-phase modulation appears near the focal point and generates distortions of the filtered spectrum. For further developments the use of a vacuum-packed setup will allow us to eliminate this effect.

Another crucial point is the evolution of the spectral phase during filtering. Unlike for the NER technique, XPW generation does not require any specific input spectral phase. Figure 2 shows the spectral phase of the pulse after the nonlinear effect, obtained from a spectral phase interferometry for direct electric-field reconstruction measurement. This measurement demonstrates that the phase of the cleaned pulse is continuous and not distorted. This phase will be easily compensated for when the filtered pulse is amplified in the second CPA setup, which can include

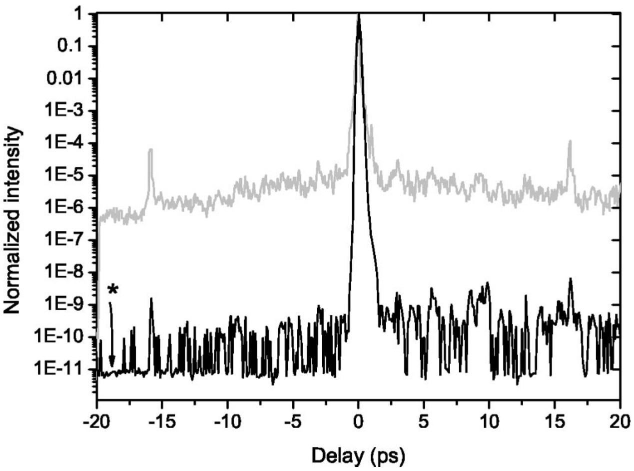

Fig. 1. Third-order correlation curves before (lighter curve) and after (darker curve) filtering. For these measurements the energy seeded in the correlator was 120 μJ. For this energy level the correlator noise was 10^{-11}.

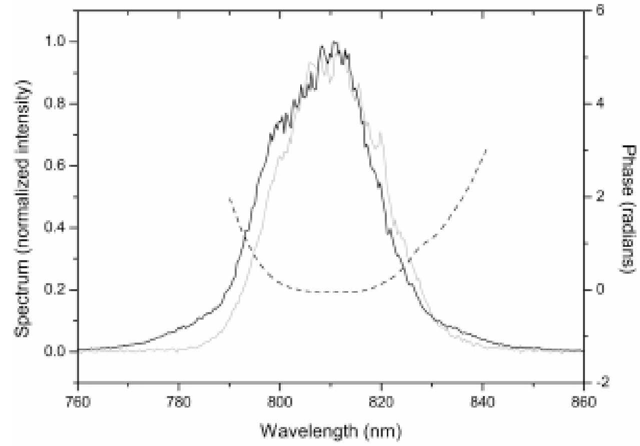

Fig. 2. Phase profile of the pulse after filtering. The input energy is 150 μJ, and the conversion efficiency is 10%.

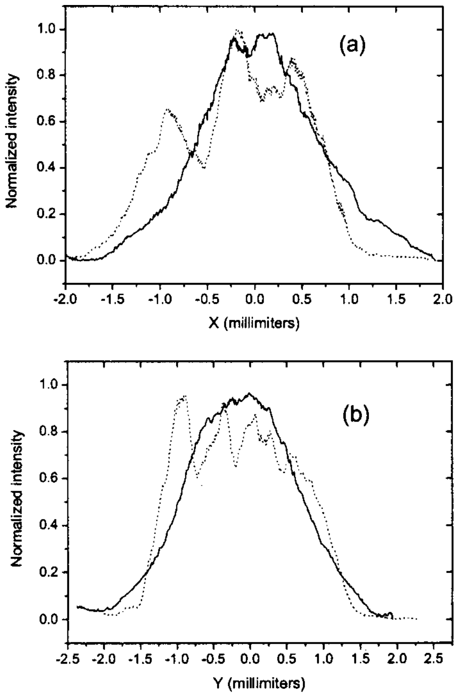

Fig. 3. (a) Horizontal and (b) vertical cuts of the spatial beam profile before (dotted curves) and after (solid curves) filtering. The input energy is 1.2 mJ, and the conversion efficiency is 10%.

a Dazzler (Fastlite).

An important characteristic for further amplification is the spatial quality of the cleaned beam, which is actually improved during the filtering. As shown in Fig. 3, even though the input spatial profile is a top hat, the filtered beam is Gaussian. Indeed, as the nonlinear effects occur in the far field, the highest spatial frequencies are removed. They are not intense enough to take part in the conversion process. So we can assert that the transmission efficiency of the filter would be improved with an input beam that has a Gaussian spatial shape and that our setup acts as an efficient temporal and spatial filter.

In conclusion, we have demonstrated a highly efficient setup for femtosecond pulse cleaning. We have designed a new kind of nonlinear filter based on cross-polarized wave generation in cubic and $\chi^{(3)}$ anisotropic crystals, such as BaF_2. Our filter is achromatic, simple, and robust, as the nonlinear process occurs in a solid medium. We achieved an energy transmission of 10%, which could be increased by use of antireflection-coated crystals. The temporal contrast is improved by more than 4 orders of magnitude, the amplified spontaneous emission intensity level is brought down to 10^{-10}, and the spatial profile is filtered. Further improvement of the temporal contrast can be achieved by careful selection of the polarizer–analyzer pair with a better extinction ratio (10^{-6}). The cleaned pulse does not exhibit any spectral phase distortions. This attractive performance allows us to believe that this technique will be useful in the design of future background-free petawatt laser systems.

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